

Serial No.: 09/544,344

PATENT APPLICATION
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AMENDMENTS TO THE CLAIMS

This listing of claims will replace all prior versions and listings of claims in the application.

1. (currently amended) An article of manufacture suitable for use in determining whether or in what amount a chemical species is present in a target environment, which article comprises
a multiplicity of particles in three-dimensional close-packed orientation, said particles having
a core of conductive metal or conductive metal alloy, in each said particle such core being of 0.8 to 40.0 nm in maximum dimension and
deposited on said core a ligand shell of thickness from 0.4 to 4.0 nm and
composed of an encapsulating monomolecular layer of ligand shell
molecules each molecule having a head-tail type structure,
the head being a functional group possessing a bonding interaction with
metal atoms in the core surface, and
the tail having a structure and composition designed to provide additional
stabilization of metal clusters against irreversible agglomeration,
induce solubility in solvents, and promote interactions with
chemical species of interest, and having a tail functional group
capable of selective interactions that discriminate between
chemical species of interest, the tail functional group selected from
the group consisting of a heterofunctional group, an aromatic
group, a secondary aliphatic group, and a tertiary aliphatic group,
the ligand shell being capable of interacting with a chemical species in a
target environment such that an electrical property of said
multiplicity of particles is altered; and
a substrate suitably configured for supporting the multiplicity of particles and presenting
the multiplicity of particles to contact with said environment.
2. (original) An article of manufacture as defined in claim 1, wherein said core comprises
silver, gold, platinum or palladium, or an alloy of two or more such metals.

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3. (canceled)
4. (currently amended): An article of manufacture as defined in claim 1, wherein said ligand shell molecule comprises a thiol or an amine in the head portion of the ligand shell molecule ~~its structure~~ as the functional group possessing a bonding interaction with metal atoms in the core surface.
5. (previously presented) An article of manufacture as defined in claim 4, wherein said ligand shell molecule comprises a primary aliphatic hydrocarbon moiety in the tail portion of its structure.
6. (currently amended) An article of manufacture as defined in claim 4, wherein said ligand shell molecule comprises an amine in the head portion ~~selected from the group consisting of primary aliphatic amines.~~
7. (original) An article of manufacture as defined in claim 1, wherein in each said particle the core is of size from 2 to 20 nm in maximum dimension and the ligand shell is of thickness from 0.4 to 2.5 nm.
8. (original) An article of manufacture as defined in claim 1, wherein the particles are substantially spherical.
9. (currently amended) An article of manufacture as defined in claim 1, wherein the ligand shell molecule ~~shell~~ contains a thiol functional group in the head portion ~~of the structure and a heterofunctional group in the tail portion of the structure capable of selective interactions that discriminate between chemical species of interest.~~
- 10-20. (canceled)

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21. (currently amended) An assembly suitable for investigation of a target environment to determine whether or in what amount a chemical species may be present, which comprises
- (a) a substrate suitably configured for presenting a multiplicity of particles supported thereon to contact with said environment;
 - (b) supported by said substrate, a film of from 5 to ~~2,000~~ 10,000 nm comprising a multiplicity of particles in three-dimensional close-packed orientation, said particles having a core of conductive metal or conductive metal alloy, in each said particle such core being of 0.8 to 40.0 nm in maximum dimension and deposited on said core a ligand shell of thickness from 0.4 to 4.0 nm and composed of an encapsulating monomolecular layer of ligand shell molecules each molecule having a head-tail type structure, the head being a functional group possessing a bonding interaction with metal atoms in the core surface, and the tail having a structure and composition designed to provide additional stabilization of metal clusters against irreversible agglomeration, induce solubility in solvents, and promote interactions with chemical species of interest, the ligand shell being capable of interacting with a chemical species in a target environment such that an electrical property of said multiplicity of particles is altered; and
 - (c) a sensor for monitoring said property of said multiplicity of particles.
22. (original) An assembly as defined in claim 21, wherein said core comprises silver, gold, platinum or palladium or an alloy of two or more of such metals.
- 23-24. (canceled)

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25. (currently amended) An assembly suitable for investigating a target environment, to determine whether or in what amount a chemical species may be present, which comprises
- (a) a substrate suitably configured for presenting a multiplicity of particles in three-dimensional close-packed orientation supported thereon to contact with said species;
 - (b) supported by said substrate, said multiplicity of particles having a core of conductive metal or conductive metal alloy, in each said particle such core being of 0.8 to 40.0 nm in maximum dimension and deposited on said core a ligand shell of thickness from 0.4 to 4.0 nm and composed of an encapsulating monomolecular layer of ligand shell molecules each molecule having a head-tail type structure, the head being a functional group possessing a bonding interaction with metal atoms in the core surface, and the tail having a structure and composition designed to provide additional stabilization of metal clusters against irreversible agglomeration, induce solubility in solvents, and promote interactions with chemical species of interest, and having a tail functional group capable of selective interactions that discriminate between chemical species of interest, the tail functional group selected from the group consisting of a heterofunctional group, an aromatic group, a secondary aliphatic group, and a tertiary aliphatic group, the ligand shell being capable of interacting with a chemical species in a target environment such that the an electrical property of the particles is altered;
 - (c) a pair of electrodes, each in electrical contact with said multiplicity of particles; and
 - (d) a sensor for monitoring the electrical property of said multiplicity of particles to determine whether there is, or the amount of, any change in said electrical property as an indication of whether or in what amount said species is present.

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26. (currently amended) An assembly as defined in claim 25, wherein
the core comprises gold and
the ligand shell molecule is selected from the group consisting of ~~primary~~
~~aliphatic thiols~~, secondary aliphatic thiols, tertiary aliphatic thiols,
heterofunctionally substituted aliphatic thiols, aromatic thiols,
heterofunctionally substituted aromatic thiols, and heterofunctionally
substituted araliphatic thiols.

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27. (currently amended) A method of fabricating an assembly suitable for investigation of a target environment to determine whether or in what amount a chemical species may be present, which comprises
- (a) depositing on a substrate
- (i) a pair of interdigitated electrodes each having a comb-like configuration and
- (ii) in such manner that the electrodes are electrically connected, a thin film of a multiplicity of particles in a three-dimensional close-packed orientation having
- a core of conductive metal or conductive metal alloy, in each said particle the core being from 0.8 to 40.0 nm in maximum dimension and deposited on said core a ligand shell of thickness from 0.4 to 4.0 nm and composed of an encapsulating monomolecular layer of ligand shell molecules each molecule having a head-tail type structure, the head being a functional group possessing a bonding interaction with metal atoms in the core surface, and the tail having a structure and composition designed to provide additional stabilization of metal clusters against irreversible agglomeration, induce solubility in solvents, and promote interactions with chemical species of interest, and having a tail functional group capable of selective interactions that discriminate between chemical species of interest, the tail functional group selected from the group consisting of a heterofunctional group, an aromatic group, a secondary aliphatic group, and a tertiary aliphatic group.
- the ligand shell being capable of interacting with a chemical species in a target environment such that an electrical property of said multiplicity of particles is altered; and
- (b) connecting said pair of electrodes with a sensor capable of determining a change in the electrical property of said multiplicity of particles.

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28. (previously presented) A method of fabrication as defined in claim 27, wherein said deposition of a thin film of the multiplicity of particles comprises spraying on the electrodes and on the substrate a solution comprising the multiplicity of particles and a solvent, said electrodes and substrate being at a temperature such that the solvent is flashed away or rapidly evaporated.
29. (currently amended) A method of fabrication as defined in claim 27, wherein said deposition of a thin film of the multiplicity of particles comprises
- (a) treating said electrodes and substrate with a difunctional material capable of binding with (i) the electrodes and the substrate and (ii) said multiplicity of said particles, such that said material binds with said electrodes and said substrate;
 - (b) contacting the treated electrodes and substrate with said multiplicity of particles ~~having a core of conductive metal or conductive metal alloy, in each said particle the core being from 0.8 to 40.0 nm in maximum dimension, and deposited on said core a ligand shell having a thickness from 0.4 to 4.0 nm and a composition as an encapsulating monomolecular layer of ligand molecules, each molecule having a head-tail type structure, the head being a functional group possessing a bonding interaction with metal atoms in the core surface, and the tail having a structure and composition designed to provide additional stabilization of metal clusters against irreversible agglomeration, induce solubility in solvents, and promote interactions with chemical species of interest, the ligand shell being capable of interacting with said species such that an electrical property of said multiplicity of particles is altered, such that said multiplicity of particles bonds with the material to form a composite comprising a layer of said particles on the electrodes and substrate.~~
30. (original) A method of fabrication as defined in claim 29, wherein the multiplicity of particles forms a monolayer on said substrate and electrodes.

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31. (currently amended) A method of fabrication as defined in claim 29, which further comprises a cycle of steps including
- (a) exposing the outwardly facing surfaces of said particles of the composite to a coupling agent capable of bonding said particles to a further multiplicity of such particles deposited thereon, ~~the particles of said further multiplicity having a core of conductive metal or conductive metal alloy, in each said particle the core being from 0.8 to 40.0 nm in maximum dimension, and deposited on said core a ligand shell having a thickness from 0.4 to 4.0 nm and of composition as an encapsulating monomolecular layer of ligand molecules each molecule having a head-tail type structure, the head being a functional group possessing a bonding interaction with metal atoms in the core surface, and the tail having a structure and composition designed to provide additional stabilization of metal clusters against irreversible agglomeration, induce solubility in solvents, and promote interactions with chemical species of interest, the ligand shell being capable of interacting with said species such that an electrical property of said multiplicity of particles is altered; and~~
- (b) contacting the particle surfaces so exposed with said further multiplicity of particles such that
- said further multiplicity of particles bonds with the particle surfaces of said composite, and
- the further multiplicity of particles is immobilized on those surfaces.
32. (original) A method of fabrication as defined in claim 31, wherein said cycle is performed a plurality of times.

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33. (currently amended) A system suitable for investigating a target environment to determine whether or in what amount a chemical species may be present, which comprises
- (a) a multiplicity of particles in three-dimensional close-packed orientation, said particles having
- a core of conductive metal or conductive metal alloy, in each said particle such core being of 0.8 to 40.0 nm in maximum dimension and deposited on said core a ligand shell of thickness from 0.4 to 4.0 nm and composed of an encapsulating monomolecular layer of ligand shell molecules each molecule having a head-tail type structure, the head being a functional group possessing a bonding interaction with metal atoms in the core surface, and the tail having a structure and composition designed to provide additional stabilization of metal clusters against irreversible agglomeration, induce solubility in solvents, and promote interactions with chemical species of interest, the ligand shell being capable of interacting with a chemical species in a target environment such that an electrical property of said multiplicity of particles is altered;
- (b) means for exposing said multiplicity of particles to said environment, said means for exposing having a substrate suitably configured for supporting the multiplicity of particles;
- (c) means for subjecting said multiplicity of particles to conditions sufficient for said property to be exhibited; and
- (d) means for monitoring said property to determine whether there is, or the amount of, any change in said [[such]] property as an indication of whether or in what amount said species is present.

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34. (currently amended) A system for investigating a target environment to determine whether or in what amount a chemical species may be present, which comprises
- (a) a multiplicity of particles in three dimensional close-packed orientation, said particles having
- a core of conductive metal or conductive metal alloy, in each said particle such core being of 0.8 to 40.0 nm in maximum dimension and
- deposited on said core a ligand shell of thickness from 0.4 to 4.0 nm and
- composed of an encapsulating monomolecular layer of ligand shell molecules each molecule having a head-tail type structure,
- the head being a functional group possessing a bonding interaction with metal atoms in the core surface, and
- the tail having a structure and composition designed to provide additional stabilization of metal clusters against irreversible agglomeration, induce solubility in solvents, and promote interactions with chemical species of interest,
- the ligand shell being capable of interacting with a chemical species in a target environment such that an electrical property of said multiplicity of particles is altered;
- (b) means for exposing said multiplicity of particles to said environment, said means for exposing having a substrate suitably configured for supporting the multiplicity of particles;
- (c) means for passing an electrical field through said multiplicity of particles; and
- (d) means for monitoring the electrical property of said multiplicity of particles to determine whether there is, or the amount of, any change in said electrical property as an indication of whether or in what amount said species is present.
35. (original) A system as defined in claim 34, wherein said means for monitoring the electrical resistivity of said multiplicity of particles includes a current-to-voltage converter circuit followed by a precision rectifier and low-pass filter.

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36. (original) A system as defined in claim 35, wherein said means further includes a voltage-to-frequency converter.
37. (currently amended) An article of manufacture as defined in claim 1 ~~claim 5~~, wherein said heterofunctional group is heterofunctionally substituted aliphatic thiol or said ~~heterofunctionally substituted araliphatic thiol is substituted by OH, COOH, NH₂, or Cl.~~
38. (currently amended) An article of manufacture as defined in claim 1 ~~claim 5~~, wherein said ~~heterofunctionally substituted aliphatic thiol~~ is further comprising a ligand molecule having the formula HS(CH₂)₆OH.
39. (canceled)
40. (currently amended) An article of manufacture as defined in claim 1, wherein the ligand shell molecule ~~shell~~ contains a thiol functional group in the head portion of the structure and
[[a]] the heterofunctional group comprising individually or a combination of an alcohol, phenol, fluoroalcohol, carboxylic acid, ether, phosphoryl, or halide heteroatom ~~heteratom~~ functional groups within the tail portion of the structure which are capable of selective interactions that discriminate between chemical species of interest.

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41. (currently amended) An article of manufacture as defined in claim 1, wherein said ligand shell molecule comprises a thiol in the head portion of the structure and comprises a secondary or tertiary aliphatic hydrocarbon structure, [[or]] an aromatic hydrocarbon structure, [[or]] an araliphatic hydrocarbon structure, [[or]] a heterofunctional aliphatic structure, [[or]] a heterofunctional aromatic structure, or a heterofunctional araliphatic structure in the tail portion of the ligand shell molecule structure.
42. (previously presented) An article of manufacture as defined in claim 41, wherein said heterofunctionality comprises a hexafluoroacetone derived adduct.
43. (previously presented) A method of fabrication as defined in claim 29, wherein said difunctional material comprises dithiol and silane thiol coupling agents.
44. (currently amended) A method of fabrication as defined in claim 31, wherein said coupling agent ~~difunctional material~~ comprises a dithiol molecule ~~coupling agent~~.
45. (currently amended) A method of fabrication as defined in claim 29 ~~claim 32~~, wherein said difunctional material comprises a dithiol molecule ~~coupling agent~~.
46. (new) An article of manufacture as defined in claim 5, wherein said ligand shell molecule comprises a C₆-C₁₂ primary aliphatic hydrocarbon moiety in the tail portion of its structure.
47. (new) An article of manufacture as defined in claim 1, wherein said ligand shell molecule is selected from the group consisting of benzyl mercaptan, phenylethyl mercaptan, araliphatic mercaptan, and 4-methoxybenzyl mercaptan.
48. (new) A method of fabrication as defined in claim 31, wherein said coupling agent is selected from the group consisting of dithiol, octanedithiol, and hexanedithiol.

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49. (new) An assembly as defined in claim 21, wherein the film is up to about 400 nm thick.
50. (new) An article of manufacture as defined in claim 1, wherein said aromatic group, said secondary aliphatic group, or said tertiary aliphatic group is substituted by OH, COOH, NH₂, or Cl.
51. (new) An article of manufacture as defined in claim 1, wherein the tail functional group is selected from the group consisting of heterofunctional group and aromatic group.